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VOLUME CHANGES IN METAMORPHISM

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Introduction.—It is the purpose of this paper to consider volume changes of rocks in relation to metamorphism. This is a subject which has already received much attention by geologists and physical chemists, but it seems to me that the former have not always appreciated the true conditions of metasomatism, while the latter have limited themselves largely to the consideration of transformations in systems in open space. The changes in the rigid rocks are subject to certain limitations to which I believe sufficient attention has not been directed.

Known changes of volume.—It is evident and well known that volume changes in rocks take place. The most efficient agencies are molecular forces: heating and cooling, fusion and solidification, molecular rearrangement (as when a mineral passes any inversion point). All of these may affect the volume of large or small masses of rocks. Injection of igneous material may increase the volume. Stretching of a geological body may result in cracks and fissures, increasing the bulk volume, and if these are filled by circulating solutions the actual volume will be greater than before. In the same way processes of filling may increase the actual volume of a porous rock. It is also well known that diminution of volume may take place in unconsolidated sediments under pressure, by closing of pores or other openings, and by incidental squeezing out of any fluid or gaseous phase which the rock may contain. In such rocks lateral movements of plastic material may occur, though this would only effect a relative change of volume. Similar relative volume changes occur by recrystallization under conditions of rock flowage (as in case of marble or ice).

Close to the surface where disintegrating agencies are at work volume changes may take place. The pressure is slight and chemical reactions proceed accompanied by increase or decrease of volume. Here hydration will result in expansion and strong solvent action may be followed by contraction of volume.

Metamorphism and metasomatism.—Metamorphism is here defined as the sum total of the chemical and mechanical changes which take place in solid rocks, below the zone of oxidation. The agents are heat, pressure, and chemical energy. Metamorphism by pressure is probably always accompanied by some chemical change. Chemical energy works by means of solutions, gaseous or liquid, which penetrate the rocks on capillary and supercapillary openings.

Metasomatism is here defined broadly as any change in composition of a mineral when exposed to conditions under which it is unstable. Solutions, gaseous or liquid, effect the change. A more restricted definition is that metasomatism comprises any change in composition of a mineral in a solid rock induced by a change in the physical conditions and resulting in its space being occupied by another mineral stable under the prevailing conditions.

Metamorphism is in most cases accompanied by metasomatism of individual minerals and usually also by metasomatism of the rock as a whole, though the latter change may proceed very slowly. If the supply of new material is rapid the composition of the rock may be greatly changed within a short time. Replacement is used as equivalent to metasomatism.

Thesis of this paper.—In a previous paper,[1] principally devoted to metasomatism in mineral deposits, I have advanced the view that replacement normally occurs without change of volume of individual minerals or rocks. I would now like to broaden this theory by the thesis that metamorphism by replacement does not normally involve changes of volume.

Replacement.—Replacement in solid rocks consists in solution of the host mineral, followed immediately by deposition of an equal volume of the guest mineral. This is an empirical observation based on the microscopic examination of rocks. In other words the volume of the replacing mineral equals the volume of the mineral replaced. Deposition follows so closely upon solution that at no

[1] "The Nature of Replacement," *Econ. Geology*, VII (1912), 521–35.

time can any open spaces be discerned under the microscope. It is evident, however, that capillary spaces are necessary for the movement of the fluid phases which in most cases are aqueous solutions. In fact, replacement usually begins from such capillary cracks or from a series of minute fluid inclusions along such a fissure. Replacement is primarily caused by solution or decomposition of the host mineral in solute films separating grains and filling capillary fissures. The solution of the host mineral produces supersaturation in the liquid, and an amount of secondary material is precipitated equal to the volume of the host mineral removed.

Replacement proceeds particle by particle but not "molecule for molecule." In other words there is no simple molecular equivalence between the material dissolved and the material precipitated. Replacement proceeds independently of specific gravity and molecular volume. Chemical reactions may take place in the contact film, but the sum total of the change is not expressible by the chemical formulas usually supposed to represent the process.

In some cases there is simply solution and deposition without chemical reaction, as when pyrite replaces calcite or limestone or shale, but whether or not the exchange is accompanied by chemical reaction does not influence the law of equal volumes. Replacement is dependent upon the velocity of the solution movement and naturally also upon composition, pressure, and temperature of the replacing solution. If the speed and chemical activity of the solutions be great the empirical volume law may fail to hold, deposition will lag behind solution, and a drusy texture will result. This happens occasionally in some metasomatic processes in the formation of mineral deposits, but in metamorphism of rocks the solutions move slowly and only a small amount of new material is introduced, so that the resulting textures are always compact. Even in the case of drusy texture the bulk volume remains the same and subsequent rock pressure fails to close the cavities, even under the weight of a rock column of 10,000 to 15,000 feet. Ultimately, at extreme depths these cavities would most probably be closed with permanent reduction of volume if they have not previously been filled by cementing solutions.

While, therefore, in extreme cases of metasomatism there may be diminution of actual volume expressed in porosity, the opposite, i.e., increase in volume, it is believed, will not take place by replacement in rigid rocks.

Form and texture of guest mineral.—The guest mineral may form aggregates with their irregular outlines simply determined by the ways open for the replacing solutions. In this case, as in replacement of feldspar by sericite, there are capillary openings between the individuals of sericite, and the space is thus filled less continuously than in the host mineral. In other words the porosity may increase by several per cent. The altered rock is then more accessible to later solutions and may easily suffer further alteration if the solutions change so as to make the new mineral unstable.

In other cases the guest mineral is compact and may assume its own crystal form, owing, it is believed, to differential pressure and corresponding difference in rapidity of growth in certain directions. At times the replacement may proceed so rapidly that parts of the host mineral or host minerals become inclosed by the guest mineral, as frequently seen in quartz crystals in limestone or garnets and ottrelites in crystalline schists.

Movement of solutions on capillary fissures.—I am aware that attempts have been made to show that circulation of solutions on capillary fissures is so slow as to be negligible.[1] While not able to refute these calculations, I can only say that the whole process of metamorphism appears to be opposed to such a conclusion.

Metasomatic shells.—It is frequently observed that metasomatic crystals, for instance, cubes of pyrite in feldspar, are surrounded by a thin covering of another mineral, i.e., quartz or chlorite following the outlines of the crystal. This is believed to mean that at a certain stage the iron solutions failed. The replacement proceeded, but instead of pyrite the next combinations available in the replacing solutions were precipitated.

Complex replacement.—One guest mineral may replace two or several host minerals without change of its crystallographic form. One pyrite crystal may extend across the contact of a quartz and a feldspar grain. In schists one garnet crystal may replace hundreds of small individuals of biotite, quartz, and feldspar.

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[1] J. Johnston and L. A. Adams, *Centralblatt für Mineralogie*, 1914, p. 171.

Examples of replacement.—The following instances are mentioned, in order to express more precisely what processes are believed to be active in certain cases.

In the simple case when pyrite replaces calcite the solutions need to carry iron sulphide, alkaline sulphides, and carbon dioxide. As each particle of calcite is dissolved as bicarbonate, the solution becomes supersaturated for FeS_2 and an equal volume of this compound is precipitated.

In the more complicated case of replacement of orthoclase by pyrite the entering solution would have the same composition as indicated above. As each particle of orthoclase is decomposed into potassium carbonate, colloid silica, and colloid aluminum silicate, all of which are carried away, a corresponding volume of FeS_2 is deposited.

When pyrite replaces chlorite the entering solution may contain only hydrogen sulphide, iron carbonate, and carbon dioxide. As each particle of chlorite is decomposed into magnesium bicarbonate, ferrous bicarbonate, colloid silica, and colloid aluminum silicate, a reaction takes place between the ferrous carbonate and the hydrogen sulphide, resulting in the precipitation of an equal volume of iron sulphide. The guest mineral contains much more iron than the host mineral and the additional amount needed must be supplied by the entering solution.

Every petrographer knows how frequently soda-lime feldspar or anorthite is replaced by calcite. It is usually explained as a simple case of the lime in the feldspar combining with carbon dioxide. A calculation based on respective specific gravities of 2.75 and 2.71 will show that one cubic centimeter of anorthite yields 2.20 grams alumina and silica, which must be carried away. In order that the resulting calcite may fill the space made vacant by the anorthite the solutions must supply an additional amount of 0.97 grams CaO besides the necessary CO_2, considering that one cubic centimeter of calcite contains 1.52 grams CaO and 1.19 grams CO_2.

The conversion of olivine (forsterite) to iron-free serpentine is usually expressed by the formula:

$$2Mg_2SiO_4+2H_2O+CO_2=H_4\,Mg_3Si_2O_9+MgCO_3.$$

Replacement by equal volumes demands that one cubic centimeter of forsterite (Sp.G. 3.21) must be decomposed and its space filled by serpentine (Sp.G. 2.50). The former measure contains 1.83 grams MgO and 1.38 grams SiO_2, while one cubic centimeter of pure serpentine contains 1.08 grams MgO, 1.10 grams SiO_2, and 0.33 grams H_2O. It is therefore necessary that 0.33 grams water should be added and that 0.75 grams MgO and 0.28 grams SiO_2 should be carried away. This indicates that the foregoing formula does not express the process correctly.

The well-known alteration of orthoclase to sericite is usually considered to be governed by one of the following formulas:

$$3KAlSi_3O_8+H_2O+CO_2=K\,HAl_3Si_3O_{12}+K_2CO_3+6SiO_2$$
$$3KAl\,Si_3O_8+H_2O=KHAl_3Si_3O_{12}+K_2SiO_3+5SiO_2$$

One cubic centimeter of orthoclase (Sp.G. 2.60) contains 1.68 grams SiO_2, 0.48 grams Al_2O_3, and 0.44 grams K_2O. One cubic centimeter of sericite holds 1.25 grams SiO_2, 1.06 grams Al_2O_3, 0.33 grams K_2O, and 0.12 grams H_2O. The replacement by equal volume, therefore, demands removal in solution of 0.43 grams SiO_2 and 0.11 grams K_2O, but also an addition of 0.58 grams Al_2O_3 and 0.12 grams H_2O. This indicates that the formulas cited above are certainly incorrect. The exact formula expressive of what has happened is probably very complicated. A crude approximation would be represented by the following equation:

$$3(K_2O\cdot Al_2O_3\cdot 6\,SiO_2)+4H_2O+3\,Al_2O_3+CO_2$$
$$=2(2\,H_2O\cdot K_2O\cdot 3\,Al_2O_3\cdot 6\,SiO_2)+6\,SiO_2+K_2CO_3$$

It is thus seen that sericitization involves a great addition of alumina. From where is this derived? It must be remembered that sericitization is usually accompanied by many other metasomatic processes involving replacement of aluminous silicates by pyrite, calcite, and chlorite. All these processes involve the liberation of much alumina which is ready to enter into the sericite molecule. The last example also shows that alumina is by no means an immobile constituent during the metamorphic processes.

Preservation of texture and structure.—The best proof that no volume change takes place during metamorphism is furnished by the frequent preservation of texture and structure. We find the sharp outlines of an olivine crystal preserved, though it may be wholly converted into serpentine. We find calcareous oölites metasomatized by fine-grained quartz; silicified dolomites similarly preserve the outlines of their individual crystals. Fossils are preserved in exact outlines after silicification or pyritization. Basic rocks are chloritized with perfect preservation of outlines of ferromagnesian silicates. Zinc carbonate sometimes reproduces faithfully the texture and structure of limestone.

Replacement under uniform pressure.—It will be necessary to consider replacement under uniform pressure separately from the more complicated conditions of stress. Uniform pressure results in increase of solubility, but the effects are slight compared to a relatively small change in temperature.[1] Uniform pressure obtains in static metamorphism, which, for instance, is active in the upper part of the crust, where hydration is an important process and the temperature not high. Rocks penetrated by solutions under static metamorphism develop many minerals by replacement, for instance, sericite, chlorite, calcite, epidote, serpentine, pyrite, etc. In many cases there would be a tendency toward increase of volume because heavier minerals are replaced by hydrated products. It is believed, however, that the law of equal volumes holds strictly in case of this form of metamorphism and that changes of volume have been more frequently assumed to exist than actually proved. Consider an ordinary diabase altered to greenstone. Such a rock is a veritable laboratory of metasomatism: sericite after feldspar, chlorite after augite, epidote, calcite, and quartz after any of the primary minerals. All these replacements are constantly going on in such a rock, and I think that in no case has any change of volume been actually proved. It would not seem impossible to do so, for instance, in case of a thick dike in sedimentary rocks.

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[1] J. Johnston and Paul Niggli, "The General Principles Underlying Metamorphic Processes," *Jour. Geol.*, XXI (1913), 504.

The serpentinization of magnesian rocks is often cited as a clear case of expansion by replacement. Recalling the olivine crystals referred to above, I believe it improbable that such an expansion has taken place. Large serpentine masses do not usually show the smooth slickensides supposed to be caused by this movement but are rather compact and solid, the slickensided fragments being confined to crushed zones or near the surface where yielding was possible. It seems much more probable that the replacement has been effected with extensive removal of magnesia, and this is supported by the prevalence of ascending magnesian waters where serpentines abound as in California.

Expansion by hydration of anhydrite I regard in the same light. If it takes place it is under exceptional conditions of light load. Many anhydrites altering to gypsum show no evidence of expansion, the veins of the latter mineral cross-cutting the crystalline structures without disturbance. Some gypsum evidently goes into solution.

Calculation of volume changes.—In view of what has been said above it must be concluded that the application of Lepsius' volume law[1] $\left(\text{Mol. vol.} = \frac{\text{mol. weight}}{\text{Sp.G.}}\right)$ to calculate volume changes in metamorphic rocks formed under uniform pressure is unwarranted and leads to totally erroneous conclusions, unless every detail of the complicated processes is known.

Contact metamorphism under uniform pressure.—In many cases contact metamorphism proceeds under uniform rock pressure. There is, however, strong gas pressure, and if its gradient is high this may appreciably affect the replacement. Contact metamorphism may take place with or without rock metasomatism. In the special case of intense metasomatism of limestone under the influence of extremely hot and concentrated gases one would think that changes of volume would occur if anywhere. And yet the field evidence as well as the microscopical evidence is strongly opposed to such a view as I have shown in the Clifton-Morenci district; other observers have come to the same conclusion.[1] A great quantity of CaO and CO_2 are carried away, but the volume of the mass remains the same; even drusy textures are rare. Accessions from the magma to form the contact metamorphic silicates have evidently balanced any shrinkage. If there were no accessions from the outside and if the silica and alumina combined with the lime to contact metamorphic silicates, considerable shrinkage would undoubtedly result provided always that opportunity were available for the gas phase to escape. In such a case the volume changes could be calculated by means of the appropriate simple chemical equations and Lepsius' volume law.[2] As a matter of fact such rocks do not show evidence of shrinkage, such as would be afforded by drusy texture, and no field evidence indicating such contraction has been brought forward. I must conclude that here, too, calculations of volume relations on the basis of Lepsius' law applied to the ordinarily used equations are worthless as indicating the actual process. It is believed that eventual tendency to contraction is equalized by additions of substance from circulating waters at the time of metamorphism.

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[1] Van Hise expresses this law as follows: "The volume of the original compound is to the volume of the compound produced directly as their molecular weights and indirectly as their specific gravities" (*U.S. Geol. Survey, Monograph 47*, p. 209).

In the case of hornfels derived by contact metamorphism from shales the mineralogical relations have been fully covered by V. M. Goldschmidt and earlier writers, but neither the microscope nor the field examination corroborate an assumption of reduction of volume. As heavy aluminum silicates have been formed it seems that contraction would take place unless counterbalanced by additions. Here also I believe that the replacing solutions, not necessarily of magmatic origin, have carried a certain amount of material into the rock and that when a heavy silicate is formed the remainder of the space will be filled by some other substance, say quartz or feldspar.

Replacement without liquid solutions.—Water solutions certainly circulate in the rocks of the upper metamorphic zones. It would be very difficult to account for metasomatism without them. It is considered possible, however, by some investigators that replacement may be effected by gaseous phases, not of magmatic origin, but simply induced by the heat of contact metamorphism in the outer zone. Goldschmidt[1] thinks so. It seems to me, however, that if this be true there would certainly be contraction evidenced in texture and possible to detect by field or microscopic methods.

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[1] W. Lindgren, *U.S. Geol. Survey, Prof. Paper 43*, 1905; F. C. Calkins, *ibid., Prof. Paper 78*, 1913, p. 132; B. S. Butler, *ibid., Prof. Paper 80*, 1913, p. 90; J. B. Umpleby, *ibid., Prof. Paper 97*, 1917, p. 71.

[2] Joseph Barrell, *U.S. Geol. Survey, Prof. Paper 57*, 1907, p. 149; also "The Physical Effects of Contact Metamorphism," *Am. Jour. Sci.*, Fourth Series, XIII (1902), 279–96.

Metamorphism under stress.—Under unequal pressure, as pointed out by Johnston and Niggli,[2] the solubility will be greatly increased, and its influence on reactions which are accompanied by the evolution of a gas is enormous. "Unequal stress will cause reactions between solids accompanied by the development of a gas phase to proceed to an extent which would be inappreciable in the absence of stress." The conditions in a rock under heavy stress are greatly complicated by the lateral movement of material by flowage (as shown by experiments on marble, ice, etc.). This action has nothing to do with replacement, but it certainly has a tendency to compress and thin the beds. No actual contraction of volume except by the closing of pores is involved in this operation.

Under heavy stress it is commonly assumed that all capillary spaces will be closed and that circulation of water and escape of volatile phases take place with the utmost difficulty. Whether this assumption is correct is not known. It is probably not true in case of limestone, which seems to be easily penetrated by liquids and gases.

Under and after conditions of stress there is often a strong tendency to development, by replacement, of heavy aluminum silicates of small molecular volume, such as staurolite, andalusite, and cyanite. This has generally been interpreted as meaning a reduction in volume, an interpretation which at first glance seems reasonable enough. Most calculations of changes of volume according to Lepsius' volume law are based on such changes.

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[1] "The relations [referring to hornfels, etc.] are valid independently of the state of the solvents, whether these are watery solutions or a melt or whether the minerals simply are separated by a space containing their gaseous phases" (*Die Kontactmetamorphose im Kristianiagebiet*, 1911, p. 122).

[2] *Op. cit.*, p. 614.

Niggli, for instance, calculates the contractions of volume of certain ottrelite schists[1] on this basis starting with a shale of assumed composition. The conditions here are very complex, and I am sure that such computations should be undertaken with the greatest caution.

Field geology has never, as far as I am aware, proved that a contraction of volume has taken place except by rock flowage. From the standpoint of microscopic investigations there is no evidence that replacement operates in a manner different from that in rocks under uniform pressure, except that under stress the crystals tend to become elongated in direction of minimum stress. In the phenomena following relaxation of pressure, such as the development of metacrysts of garnet and andalusite, chiastolite and sillimanite, we see all the familiar phases of normal replacement, i.e., crystal form, inclusions of matrix, and complete filling of space. No crystalline schist has a drusy texture or structure indicating contraction cracks. These metacrysts or porphyroblasts develop normally across the matrix[2] and present no indication that the "force of crystallization" has bulged the rock mass, a most improbable hypothesis considering the conditions in the deep zones. Only if we assume that the rocks be actually soft is any such view tenable. It is true that the lamellae of mica sometimes bend around the crystal. These phenomena have been studied by several authors[3] who have concluded that they are caused by later mylonitic movements on gliding planes producing rotation and also development of "quartz tails" on both sides of the crystal. That rotational movements actually have occurred has been proved among others by F. H. Lahee and H. Backlund. Conditions of stress resumed after the compact crystals had been formed by replacement would naturally tend to bend the elastic mica plates around the hard bodies just as glacial clays often form layers conforming to the outline of included bowlders.

[1] *Beiträge zur geol. Karte d. Schweitz*, N.F., XXXVI, 1912.

[2] H. Rosenbusch, *Elemente der Gesteinslehre*, 1901, Figs. 73 and 74; C. K. Leith and W. J. Mead, *Metamorphic Geology*, 1915, Fig. 10.

[3] Paul Niggli, *Beiträge zur geol. Karte d. Schweitz*, N.F., XXXVI 1912; F. H. Lahee, "Crystalloblastic Order," etc., *Jour. Geol.* (1914), 500–515; H. Backlund, *Geol. För. För. Stockholm*, XL (1918), 101–203.

As the matrix is usually made up of alternating lamellae of mica and quartz it is possible that the quartz is dissolved in the direction of stress and deposited perpendicularly to it, while the mica is stable under prevailing conditions. This explanation might apply to some cases where the garnets have the appearance of having grown by forcing the schist apart. Anyway, such an action would involve an increase in volume while certainly the whole tendency in crystalline schists is more toward compression than expansion. The microscopic evidence, therefore, favors the view that replacement in crystalline schists has taken place by equal volumes, though the possibility cannot be denied that under heaviest stress the mode of replacement may be so altered that a smaller volume results.

The occurrence in many of these rocks of abundant crystals of heavy aluminum silicates raises the question whence came this concentration. The alumina could not have been derived only from the replaced host minerals; it must have been concentrated from material some distance away. It seems then that it would be necessary to assume the presence of moving liquid or gaseous solutions. And if moving solutions are admitted there is no essential difference between replacement in this case and in that of static metamorphism. These solutions would have carried other material from the outside, and the difference in volume between the newly formed heavy minerals and the dissolved host minerals would be equalized by new deposits whereby the demand for replacement by equal volume could be satisfied.

It would seem then that the conception of metamorphism under stress here outlined involves somewhat free circulation and limited addition and subtraction of material, with strong tendency toward the preservation of volume.

This is to some degree in opposition to the views of Rosenbusch and Grubenmann but in line with a recent paper by Leith and Mead,[1] in which the convergence to mineral type in dynamic metamorphism is emphasized. Duparc and others have shown us that the conversion of hornblende to uralite is not a simple paramorphism but a metasomatic process. Leith and Mead point out that the composition of a schist tends to approach the distinctive chemical characteristics of the dominant platy or columnar mineral, be it muscovite, chlorite, or amphibole. This is clearly the result of a slow metasomatic action, though this view is not definitely expressed by the authors, and if true indicates that gradual exchange of material cannot be excluded for the crystalline schists.

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[1] C. K. Leith and W. J. Mead, "Metamorphic Studies," *Jour. Geol.*, XXIII (1915), 600–607.

It is well to recognize that recrystallization in rocks under stress remains a subject beset with many difficulties and uncertainties. Fully conscious of this I advance the foregoing suggestions simply as a tentative effort from the viewpoint of a student of metasomatic action.